PB86-176146

Historic Emissions of Sulfur and Nitrogen Oxides in the United States from 1900 to 1980 (Journal Version)

Pacific Environmental Services, Inc., Durham, NC

Prepared for

Environmental Protection Agency
Research Triangle Park, NC

1986

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. PB 86-176146
EPA/600/J-86/006		

4. TITLE AND SUBTITLE	5. REPORT DATE
Historic Emissions of Sulfur and Nitrogen Oxides in the United States from 1900 to 1980 (Journal Version)	1986
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) G. Gschwandtner, K. Gschwandtner, and K. Eldridge (Pacific Environmental); C. Mann (EPA/OAQPS); and D. Mobley (EPA/AEERL)	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Pacific Environmental Services 1905 Chapel Hill Road Durham, North Carolina 27707	10. PROGRAM ELEMENT NO.
	11. CONTRACT/GRANT NO. 68-02-3887, Task 11
12. SPONSORING AGENCY NAME AND ADDRESS EPA, Office of Research and Development Air and Energy Engineering Research Laboratory Research Triangle Park, NC 27711	13. TYPE OF REPORT AND PERIOD COVERED Journal Article; September 85
	14. SPONSORING AGENCY CODE EPA/600/13

15. SUPPLEMENTARY NOTES AEERL project officer is J. David Mobley, Mail Drop 61, 919/541-2612. REF: JAPCA,36(2):139-149, February 1986

16. ABSTRACT The paper gives estimates of historic emissions of SOx and NOx for each state of the coterminous U.S. The emissions were estimates by individual source category on the state level from 1900 to 1980 for every fifth year and for 1978. The source categories included power plants, industrial boilers, industrial processes, commercial and residential heaters, natural gas pipelines, highway vehicles, off-highway diesel engines, and all other anthropogenic sources. These emissions were calculated from statistics indicative of fuel consumption or industrial output, estimations of average statewide fuel properties, and estimations of emission factors specific to each source category over time. The emission estimates were then aggregated to show the emission trends on a state, regional, and national scale. The trends are presented by source category and by major fuel type. The emission estimates allow temporal and regional trend comparisons between the emissions and chemical and biological effects trends derived in other studies. They provide a basis for assessing pollutant damage, for studying trends in stream chemistry and deposition monitoring data, and for evaluating proposed mitigation and control strategies.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution	Pollution Control	13B
Sulfur Oxides	Stationary Sources	07B
Nitrogen Oxides	Historic Emissions	
Trends		12A
History		05D

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
Release to Public	Unclassified	12
	20. SECURITY CLASS (This page) Unclassified	22. PRICE

EPA Form 2220-1 (9-73)

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA. 22161

EPA/600/J-86/006
JOURNAL ARTICLE

Historic Emissions of Sulfur and Nitrogen Oxides in the United States from 1900 to 1980

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Historic emissions of sulfur oxides and nitrogen oxides were estimated for each state of the coterminous United States. The emissions were estimated by individual source category on the state level from 1900 to 1980 for every fifth year and for 1978. The source categories included power plants, industrial boilers, industrial processes, commercial and residential heaters, natural gas pipelines, highway vehicles, off-highway diesel engines, and all other anthropogenic sources. These emissions were calculated from salient statistics indicative of fuel consumption or industrial output, estimations of average statewide fuel properties, and estimations of emission factors specific to each source category over time. The emission estimates were then aggregated to show the emission trends by state, region, and all states combined. This paper summarizes the historic emission trends on a state, regional, and national scale. The trends are presented by source category and by major fuel type. The emission estimates allow temporal and regional trend comparisons between the emissions and chemical and biological effects trends derived in other studies. They provide a basis for assessing pollutant damage, for studying trends in stream chemistry and deposition monitoring data, and for evaluating proposed mitigation and control strategies.

Sulfur oxides (SO_x) and nitrogen oxides (NO_x) are considered primary precursors of acidic precipitation. The anthropogenic emissions of these pollutants are suspected causes of many biological and chemical effects observed in recent years. Understanding the historic emission trends is important to understanding the development of acid precipitation related problems and causes of observed environmental effects.

Annual quantities of emissions of SO_x and NO_x are presented for each of the contiguous 48 states and the District of Columbia. The emissions were estimated for every fifth year from 1900 to 1980 and for 1978. The time span from 1900 through 1980 was selected to allow study of early alkalinity measurements and also to allow comparison with the 1980 national emission inventory being developed under the National Acid Precipitation Assessment Program. Five-year intervals were selected to provide an indication of the emission trends sufficient for most effects studies and to develop a methodology that could be applied to all other years. The state level was selected because it provides the most complete and consistent body of information on a historical basis and collectively covers all geographic regions of the country.

These emission estimates are improvements over previous estimates reported by Gschwandtner *et al.*[1] and included in assessment documents.[2,3] They are the result of refinement in methodology and further research. The original data file included only 33 eastern states and the District of Columbia and was limited to the time period between 1950 and 1978. This earlier file provided the best estimates of the emission trends for these states available at the time, but it was recognized that the estimates for 1950 and 1955 were incomplete and inconsistent with later years and that further improvements in the methodology were needed. The present report incorporates many techical improvements, additional details, and an expansion of the data file in both time and geographic coverage.

The state-level emission estimates for point and area sources contained in this report, supplement the national historic emission estimates of the U.S. Environmental Protection Agency

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(EPA).[4] The EPA reports total emissions of each pollutant by major source category for every tenth year from 1940 to 1970 and yearly since 1970. The state-level emission estimates in this report provide greater spatial resolution for the study of acid precipitation trends over a longer time period. These latest estimates also incorporate some information provided by other researchers concerned with historic emissions. Information on emission trends for residential sources and highway vehicles inferred by Lipfert *et al.* have been incorporated.[5] For years prior to 1950, estimates of state coal consumption estimated by Husar *et al.* have also been included in the data file.[6-8] Husar and Holloway used regional, state, and sub-state level coal consumption data to indicate the emission trends of SO_x in the early part of the century and late 1800s.[9]

This paper summarizes the general methodology and the results of the current historic emissions data file available for each pollutant. Sulfur oxides are expressed as sulfur dioxide (SO_2) and NO_x are expressed as the equivalent nitrogen dioxide to be consistent with the emissions reported by EPA. Specific methods of data reduction for each source category are described in a separate report,[10] which also compares the estimates with other emission inventories and discusses ways to further improve the estimates. The report contains the historic SO_2 and NO_x emission data file for each state by source category and the underlying fuel consumption, emission factors, and sulfur content data bases.

Method

Average emission rates for each study year were calculated for individual source categories for each state. The source categories are listed in Table I according to the fuel consumed. These categories represent all types of boilers, furnaces, engines, processes, and other anthropogenic emission sources. The basic steps involved in calculating state emissions are:

1. Obtain state level information on fuel use.
2. Allocate fuel quantity used by each source category.
3. Develop source category emission factors.
4. Determine fuel sulfur content by state for each category.
5. Calculate emissions, after emission controls.

The actual procedure varied somewhat depending on the usefulness and availability of information. It can generally be described in more detail for two time periods: 1) 1950 to 1980, and 2) 1900 to 1945.

Approach for 1950 to 1980

Fuel Consumption. For electric utilities, state consumption rates of fossil fuels were derived by individual power plant from the Bureau of Census,[11] the American Petroleum Institute,[12] the U.S. Department of Energy reports,[13] and the National Coal Association.[14] The consumption rates were determined according to boiler type. For all other fuel burning categories listed in Table I, annual fuel consumption rates were obtained from various government publications. The source of information for each category is presented by Gschwandtner *et al.*[10] When fuel consumption data were not available other salient statistics such as fuel sales, demand, distribution, and shipments were used. For the highway vehicles category, vehicle miles traveled were used for 1970 to 1980 because these provided a better estimate according to the mix of vehicle types. For earlier years, gasoline consumption by state was used. For the wildfire category, total forest area burned in each state was used.[15] For smelters and miscellaneous industrial processes, estimates were based on either individual plant or state production rates. For 1950 to 1980, state-level fuel consumption data were available for nearly all source categories.

Emission Factors. The state-level data were then multiplied by specially derived emission factors to yield estimates of uncontrolled emissions. First, the most recent emission factors of EPA, reported in AP-42 and EPA-400/9-78-005, were obtained.[16,17] These factors are based on actual emission tests of each type of combustion process or emission source represented in each source category. They are most appropriate when applied to a large number of sources such as on the state level. Periodically, the mobile and stationary source emission factors are revised by the Agency to include new, additional, or improved test data. The emission factors were held constant over time for all categories except the commercial-residential category, where the factors varied to reflect the growth of commercial sources.

The factors for each pollutant were then adjusted to represent each source category as a whole. This procedure involved mathematically weighting each factor according to the amount of fuel consumed in recent years by various types of boilers, furnaces, engines, processes, or other emission sources comprising the category. For highway vehicles, NO_x emission factors were state-specific and were weighted according to the amount of urban and rural traffic, average elevation, vehicle mix, and pollution controls in use. These adjustments provided the most representative factors for 1970 to 1980 for which vehicle-miles traveled were available. For earlier years, vehicle-miles traveled are not available, and the factors were based instead on gasoline consumption and on estimated average miles per gallon for both urban and ru-

Table I. Fuel types and emission source categories.

Bituminous coal:	Electric utilities
	Industrial boilers and space heaters
	Commercial and residential users
	Steam railroads
	Coke plants
Anthracite coal:	All uses
Residual oil:	Electric utilities
	Industrial boilers and space heaters
	Commercial and residential users
	Vessels
Distillate oil:	Electric utilities
	Industrial boilers and space heaters
	Commercial and residential heating
	Railroads
	Vessels
Natural gas:	Electric utilities
	Industrial boilers and space heaters
	Pipeline compression stations
	Commercial and residential uses
Wood:	Electric utilities
	Industrial boilers and space heaters
	Commercial heating
	Residential wood stoves and fireplaces
Gasoline and diesel fuel:	Highway vehicles
	Off-highway vehicles
	Vessels
Other:	Wildfires
	Cement plants
	Copper, lead, and zinc smelters
	Miscellaneous industrial processes
	Miscellaneous other sources

ral traffic. These factors yield generally the same result as that derived by the U.S. Department of Energy from the trend in internal compression ratios of vehicle motors.[18]

SO_2 emission factors were also weighted according to fuel consumption by individual emission sources within each category. However, these factors are more dependent on fuel properties than on combustion sources and include a fuel sulfur content variable. The emission factors account for the fraction of the fuel sulfur that would be emitted as uncontrolled emissions. The factors are determined on the basis of source emission tests and materials balance analysis involving various coal ranks that are commonly used by each source category. The average statewide values of sulfur content of coal at the consumer level were obtained from the Bureau of Mines for 1965.[19] For the earlier years, average statewide values were calculated from fuel distribution reports and information on fuel properties by originating district.[20,21] The average values of sulfur content of fuel oils were obtained from information published by the Bureau of Mines and the U.S. Department of Energy's Bartlesville Technology Center for domestically produced fuel oils.[22]

Emission Controls. The amount of emissions controlled by certain control devices was then subtracted from each source category. This step pertained to SO_2 which is largely controlled by flue gas desulfurization systems at power plants and by-product sulfuric acid plants at smelters. Controls applied to sources of NO_x emissions have generally had little effect in reducing emissions through 1980.[4]

Approach for 1900 to 1945

For this time period, state-level data on actual fuel consumption by source category were not always available, especially for the earlier years. Also, the method for collecting and reporting early data was not always consistent with the method for recent years. Depending on the type of information found, one of three approaches was taken:

1. State-level data were used when available,
2. National data were apportioned to the states, or
3. No estimates were made when state and national data were unavailable and when the emissions were so small as to be considered negligible.

These approaches help account for most of the early SO_2 emissions which were dominated by coal usage and for which consumption data are available either on the state or national level. Most NO_x emissions are also accounted for by this approach, but in terms of quantity are comparatively less than SO_2 because of the low consumption rate of fuel oils and natural gas in the early years.

Assumptions

The same sulfur content values derived for 1955 were assumed to apply to the earlier years. This assumption was made because no evidence was found to suggest a general trend in sulfur content. Available information indicates that sulfur content of coal as mined did not change significantly, and most coal was consumed in or near the producing states. Analysis of coal distribution patterns also suggests little change compared to the changes in the middle or recent part of the century.

It was also assumed that the emission factors used for 1955 applied to earlier years. No evidence was found to suggest a change in either the emission characteristics of coal-fired sources or the population mix of types of boilers and furnaces. As research in historic emission patterns and trends continues, this general assumption may be replaced by specific state-level data if such data can be developed. Other assumptions more specific to certain years and source categories are described by Gschwandtner *et al.*[10]

Aggregation of Emissions

Historic fuel consumption data were tabulated for each state according to source category and study year. Corresponding fuel sulfur content values were also tabulated for each source category according to state and year. A third tabulation contained the weighted emission factors for SO_2 and NO_x by source category and in some cases by state. These three matrices were multiplied to produce two new matrices, one for SO_2 emissions and one for NO_x emissions. The emissions of each state were then totaled by year to provide an estimate of overall national emission trends. They were also totaled by fuel type and source category to show the relative contribution of each to the overall trend. State emissions were also aggregated to show the trends in broad geographic regions of the country.

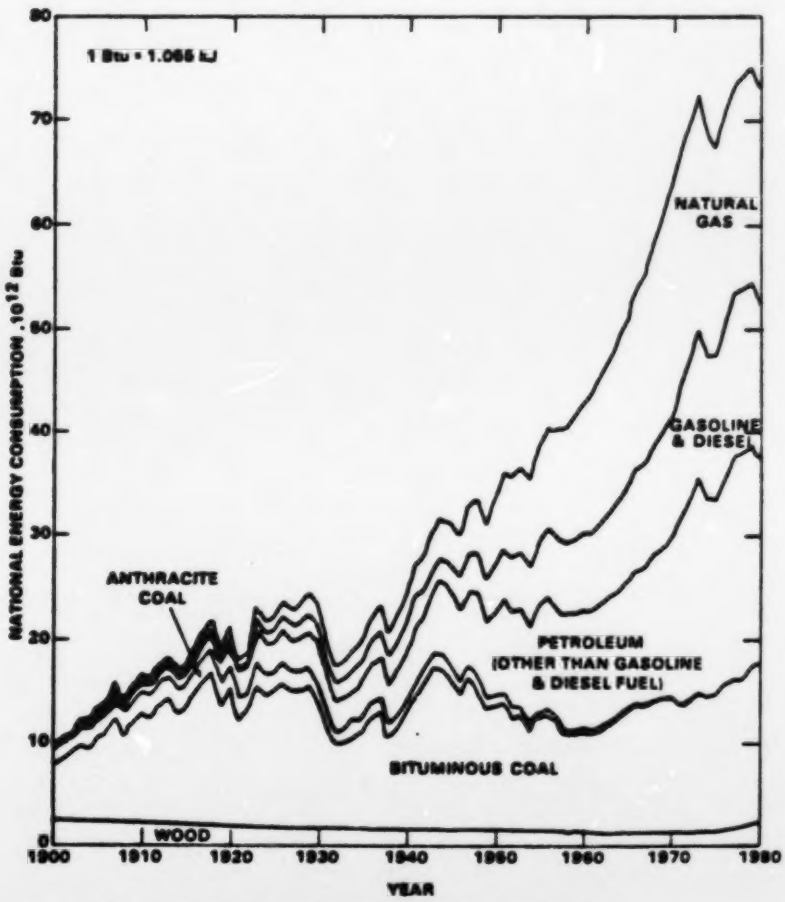

Figure 1. Total fuel consumption of the United States by fuel type: 1900 to 1980.

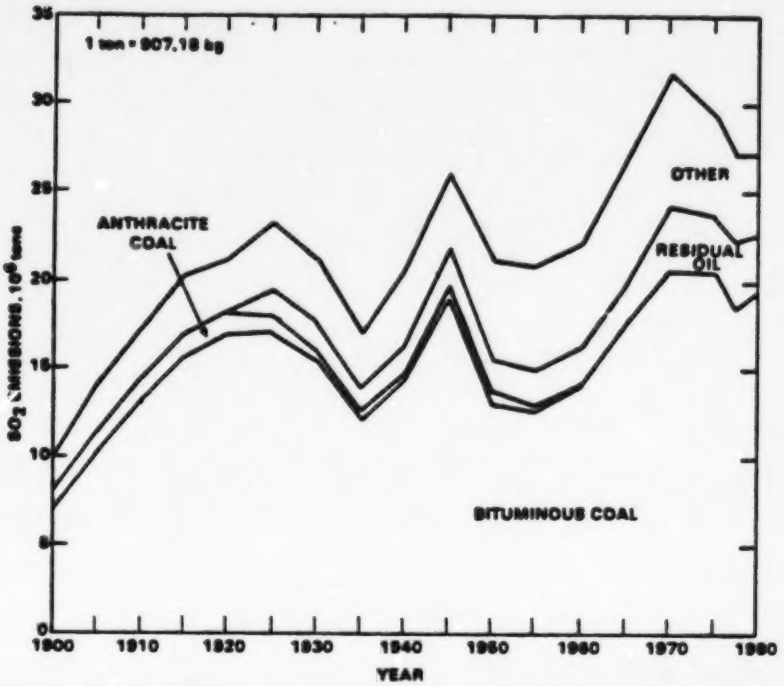

Figure 2. Overall trend in SO_2 emissions from 1900 to 1980 for the United States and by fuel type for each study year

Analysis by Release (Stack) Height

The percentage distribution of national emissions was estimated for each source category according to four broad ranges of stack heights. In the case of electric utilities, individual power plant emission estimates and stack height data were used to determine the national distribution by height from 1950 to 1980. For earlier years and for other sources, a general trend in the percentage of emissions by stack height range was estimated based largely on engineering judgement. It was assumed that the percentage emissions of each category changed linearly over time. The percentages estimated for each year were then multiplied by the total emissions for each category. However, these results should be considered less reliable than the other estimates and provide only a general indication of the national trend.

Analysis by Season

The seasonal emissions were also estimated for major source categories. These estimates were based on monthly fuel data available for only certain years, and an assumed percentage distribution of the emissions by season over time. The distribution for each category was weighted by the historic emission estimates to yield an average seasonal distribution for each study year for all categories combined.

Results for 1900 to 1980

National and regional trends of each pollutant are presented here by fuel and by source category. The trend in total emissions of individual states is also shown here.

Fuel Consumption, Overall and for Categories

Figure 1 shows the total mineral fuel consumption for the United States in terms of energy and by major source as obtained from the Bureau of Census.[23,24] In 1980, coal consumption was about the same as during the mid 1920s and 40s. Since about 1960, annual national coal consumption has increased by about 30 percent. Petroleum and natural gas consumption (the primary sources of NO_x emissions) has increased most rapidly since the 1930s. Wood and anthracite accounted for a large portion of the total energy consumption in the early part of the century but not in the latter part. Until 1930, per capita energy consumption remained nearly constant, declined during the Great Depression, and has increased since, except during the oil shortage in the mid-1970s.

SO_2 Emissions, Overall and for Categories

Figure 2 shows a plot of the total quantity of SO_2 emissions for the contiguous United States by fuel type and for each study year. (It should be noted that the emission levels for consecutive study years are interconnected only to highlight the historic trend. They do not necessarily represent the trends in the intervals between study years.) Overall, the SO_2 trend follows the general trend of coal consumption except that total emissions appear to have decreased by 10 percent from a maximum

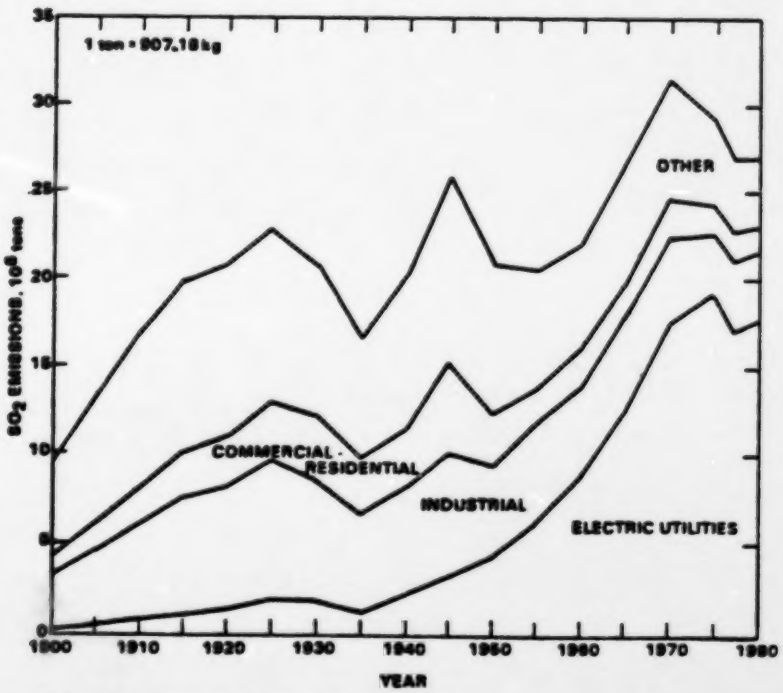

Figure 3. Overall trend in SO_2 emissions from 1900 to 1980 for the United States and by source category for each study year.

around 1970. This decrease is due largely to the general decrease in sulfur content of fuels combusted and emission reductions brought about by national and state environmental control regulations.[25] Sulfur content has decreased to a large extent as a result of coal cleaning and mixing eastern coal with cleaner western coal, while in the early years coal was mostly burned as received from the nearest coal-producing district.

Figure 3 shows the same overall trend but by source category. This plot reflects the growth of major fuel-consuming sectors and changes in fuel demand. For example, electric utility emissions appear to have increased sharply by the 1950s and 60s. In contrast, SO_2 emissions from steam locomotives almost completely disappeared by 1950 with the advent of diesel-powered railroads.

NO_x Emissions, Overall and for Categories

In sharp contrast to SO_2, total NO_x emissions appear to have increased constantly throughout most of the study period as shown in Figure 4. The total quantity of emissions is plotted on the same scale as SO_2 to allow a direct comparison. This upward trend is primarily a result of increased use of oil and natural gas. Figure 5 shows that the increase is largely due to the growth in the number of highway vehicles, natural-gas-fired power plants, and many other factors related to population growth and technology changes.

Analysis by Region

The total state emissions were aggregated according to the Administrative Regions of the EPA shown in Figure 6. These regions represent various broad geographic regions of the country. It should be noted that the regions vary in size and in the number of states and that these two factors will also affect the total regional emissions. By selecting a combination of states other than these federal regions, different emission trends may be shown. Recognizing this fact, the regions were selected to provide only a general indication of trends in various regions of the country.

The overall trend of SO_2 emissions by source category is plotted in Figure 7 for each region. These plots provide resolution of the national trend and allow the historic emission trends of each region to be compared. For example, Regions 3, 4, and 5 appear to have historically emitted more SO_2 from all categories combined than all other regions. The total SO_2 emissions of Regions 1 and 2 combined have historically remained constant. Figure 8 shows the regional trends of NO_x

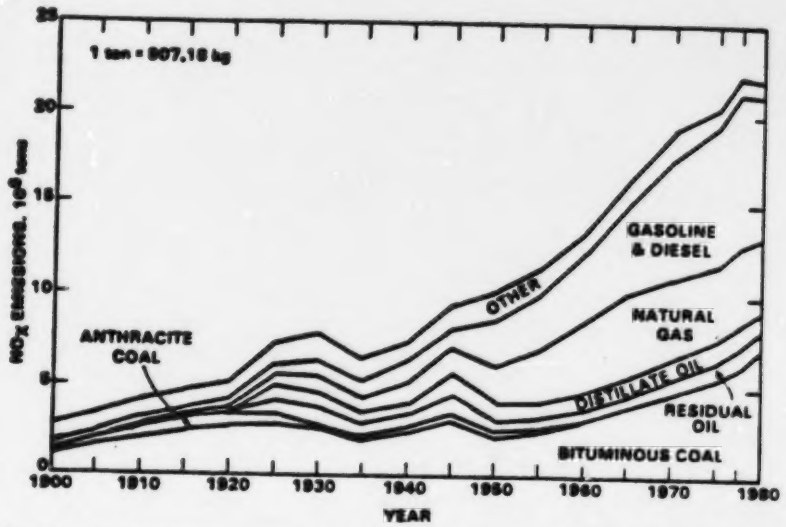

Figure 4. Overall trend in NO_x emissions from 1900 to 1980 for the United States and by fuel type for each study year.

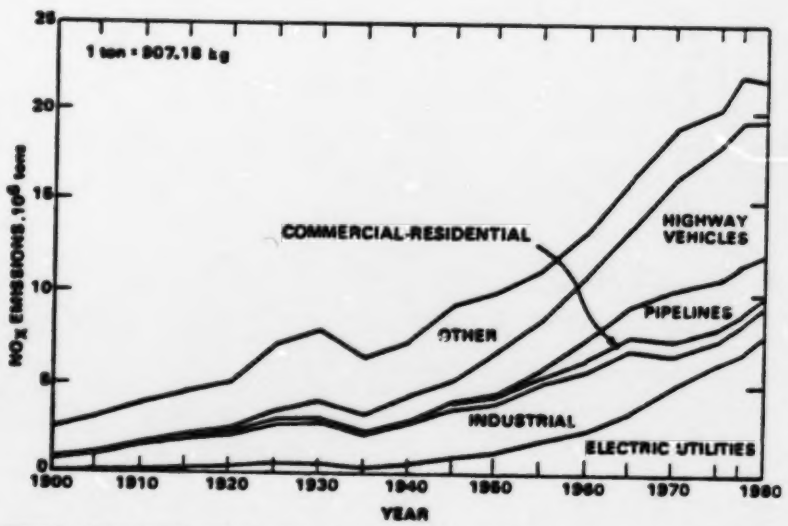

Figure 5. Overall trend in NO_x emissions from 1900 to 1980 for the United States and by source category for each study year.

Figure 6. EPA's administrative regions.

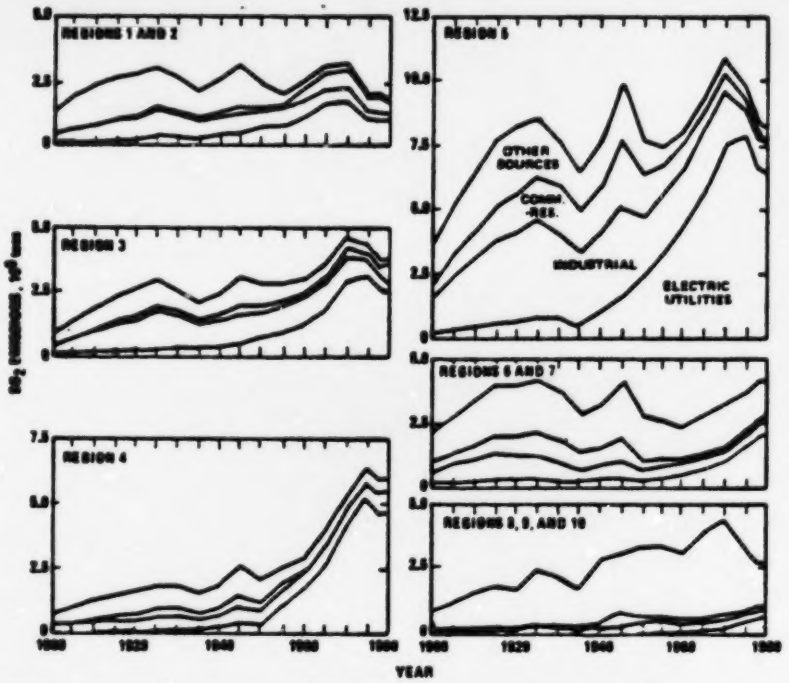

Figure 7. Temporal changes in regional SO_2 emissions by source category, 1900–1980.

emissions. In Region 6, NO_x emissions have increased more rapidly than in any other region due to the increased use of natural gas by industry and natural gas production and distribution activities. In all other regions, highway vehicles and electric utilities have accounted for more than half the total NO_x in the past several decades.

The category marked "other sources" includes railroads, smelters, wildfires, vessels, off-highway diesels, and miscellaneous other sources. Railroads were a major source of both pollutants in the early years especially in the east and midwest. Copper smelters have historically been major sources in the west.

In 1950, the regions east of the Mississippi River (Regions 1 through 5) emitted 71 percent of the total national SO_2 emissions and 60 percent of the total NO_x emissions. By 1980, these eastern regions emitted 74 percent of the total SO_2 and 57 percent of the total NO_x. During this time period, total national SO_2 emissions increased approximately 28 percent while total NO_x emissions increased 116 percent. While most of the emissions have historically originated in the east, the western regions have begun to emit a greater share of the total national NO_x emissions, especially in recent years.

Analysis by State

Figure 9 shows graphically the total emission level of each state for every tenth year since 1900. The totals were obtained by aggregating the emissions of all source categories on the state level. The figure provides a general indication of the emission trend in each state and is useful for comparing one state to another. Generally, it shows that SO_2 emissions have historically been the greatest in the northeast, especially in the heavily industrialized states. Total SO_2 emissions have historically remained highest in the industralized areas of the northeast, but in recent years have significantly increased in other states, especially those in the southeast and mid-Atlantic region. NO_x emissions have historically increased in all states, especially in those undergoing a rapid economic and population growth. The emission estimates for each source category comprising the total state emissions are contained in Volume II of Reference 10.

Distribution by Release (Stack) Height

Analysis of emissions by release height is important in studying the potential for long-range transport. Figure 10 shows the overall national trend of SO_2 and NO_x emissions by stack height range. It should be noted that the *potential* for long-range transport increases with each higher range. This analysis does not include stack exit velocities or atmospheric mixing heights, which are also important considerations. The analysis in this study suggests that more SO_2 emissions were released into the atmosphere from stacks taller than 240 ft (1 ft = 0.3048 m) than from stacks shorter than this height since about 1945. By 1980, approximately 30 percent of the SO_2 emissions were emitted above 480 ft, for example, compared to only 5 percent above this height in 1950. Not only have the percentages increased, but total national SO_2 emissions also increased and peaked around 1970. The percentage of the total SO_2 emissions released below 120 ft has generally decreased over the study period. The distribution of NO_x emissions has historically remained constant, although on the national lev-

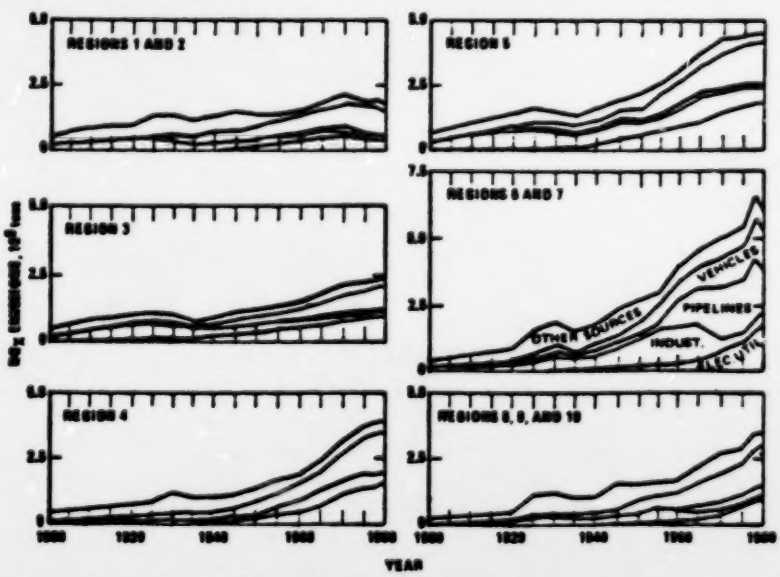

Figure 8. Temporal changes in regional NO_x emissions by source category, 1900–1980.

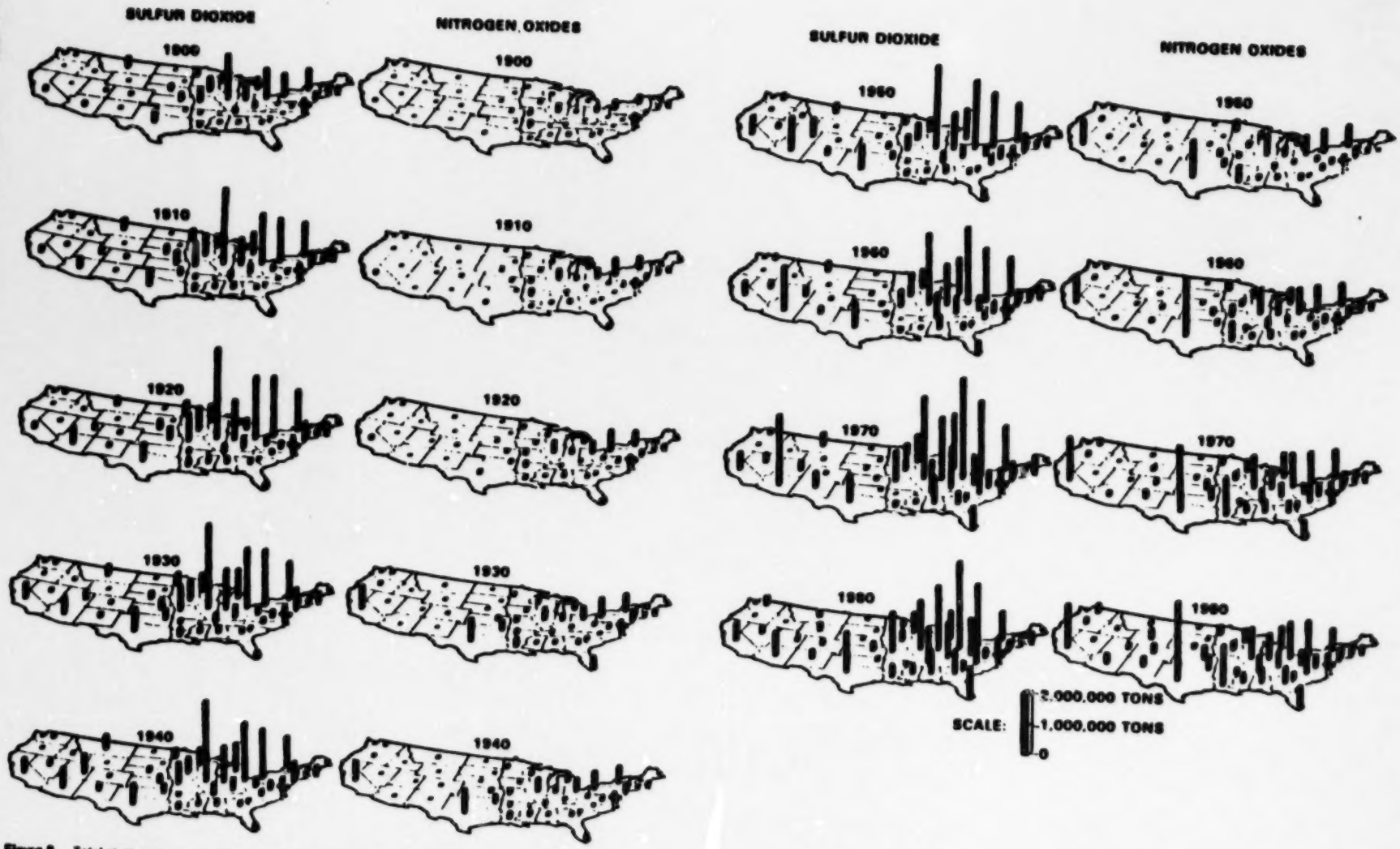

Figure 9. Total state emissions of SO_2 and NO_x for every tenth year from 1900 to 1980.

el the total emissions have steadily increased. Aproximately 60 percent of the total NO_x emissions in 1980 were released from ground level sources; predominantly from transportation sources.

Analysis of the electric utility category alone suggests that, in the 1950s and 60s, most of the SO_2 and NO_x emissions from this category were released below 480 ft—mostly between 240 and 480 ft. By 1980, about 50 percent of the total SO_2 emissions and 40 percent of the NO_x emissions from this source category were released above 480 ft as a result of the trend toward taller stacks. Since the emissions from electric utilities constitute a large portion of the total national emissions in recent years as shown in previous figures, they have a significant effect on the overall distribution of emissions by release height.

Distribution by Season

Table II presents the estimated percentage distribution of emissions by season on the national level. The percentage of wintertime SO_2 emissions appears to have decreased from about 35 percent of the total annual emissions in 1900 to about 26 percent of the total in 1980. This gradual decrease is largely a result of switching from coal to fuel oil and gas for heating, and also an increase in summertime fuel combustion by power plants to meet the demands of refrigeration and air conditioning. The seasonal distribution is more pronounced on a regional level where climatic conditions deviate from the national average. NO_x emissions appear more evenly distributed by season than SO_2 emissions. This is primarily due to an assumed even distribution of highway vehicle emissions by season, and the fact that the major sources of SO_2 emit less NO_x.

Figure 11 shows the trend in the quantity of emissions by season on the national level. While the SO_2 percentage distribution shows a gradual change, the increase in the quantity of SO_2 emissions is more evident, especially in the summertime.

Discussion

This paper has described the basic approach used to develop historic emission estimates for SO_2 and NO_x on the state level. The emissions were derived for individual source categories and groups of categories which collectively account for all anthropogenic emissions. It has presented the trends in total emissions for the contiguous United States, for sub-regions, and for individual states. The spatial and temporal trends were shown in five-year intervals. The emission trends of both pollutants vary according to the quan-

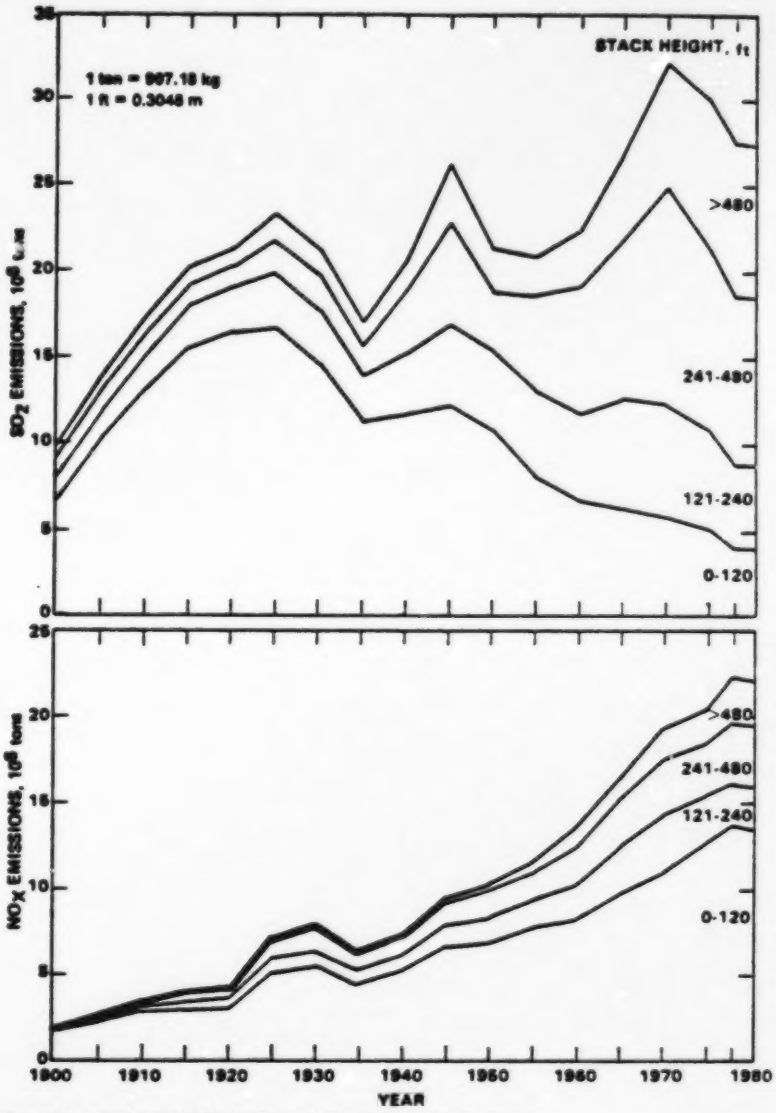

Figure 10. Total national SO_2 and NO_x emissions by stack height ranges.

Table II. Percentage distribution of emissions by season.*

Year	Percentage SO_2 emissions				Percentage NO_x emissions			
	Winter	Spring	Summer	Fall	Winter	Spring	Summer	Fall
1900	34.8	19.4	23.6	22.2	30.8	19.9	26.1	23.2
1905	34.5	19.4	24.1	22.0	31.1	19.8	26.9	23.0
1910	34.3	19.4	24.5	21.9	31.7	19.8	25.8	22.6
1915	34.3	19.3	24.7	21.7	31.9	19.8	25.8	22.4
1920	34.2	19.4	24.8	21.6	31.6	20.2	25.8	22.4
1925	34.0	19.5	25.0	21.4	30.3	20.4	26.5	22.7
1930	34.0	19.4	25.4	21.2	29.5	20.7	27.0	22.9
1935	33.4	19.4	26.0	21.2	28.8	20.9	27.4	22.9
1940	32.5	19.8	26.3	21.3	28.8	21.2	27.3	22.7
1945	32.3	20.0	25.4	21.3	28.5	21.2	27.5	22.8
1950	30.8	21.0	26.3	21.9	27.6	22.3	27.0	23.0
1955	30.2	21.9	25.7	22.2	27.2	23.0	26.7	23.2
1960	29.5	22.7	25.4	22.5	26.5	23.4	26.5	23.5
1965	28.0	23.3	25.6	23.1	25.7	23.9	26.6	23.9
1970	26.7	23.5	25.8	23.9	24.6	24.1	27.2	24.2
1975	26.1	23.5	26.1	24.2	24.2	24.1	27.3	24.3
1978	26.4	23.4	26.1	24.1	24.5	24.0	27.2	24.3
1980	26.4	23.4	26.1	24.1	24.5	24.0	27.2	24.3

* Winter = December, January, February; Spring = March, April, May; Summer = June, July, August; Fall = September, October, November.

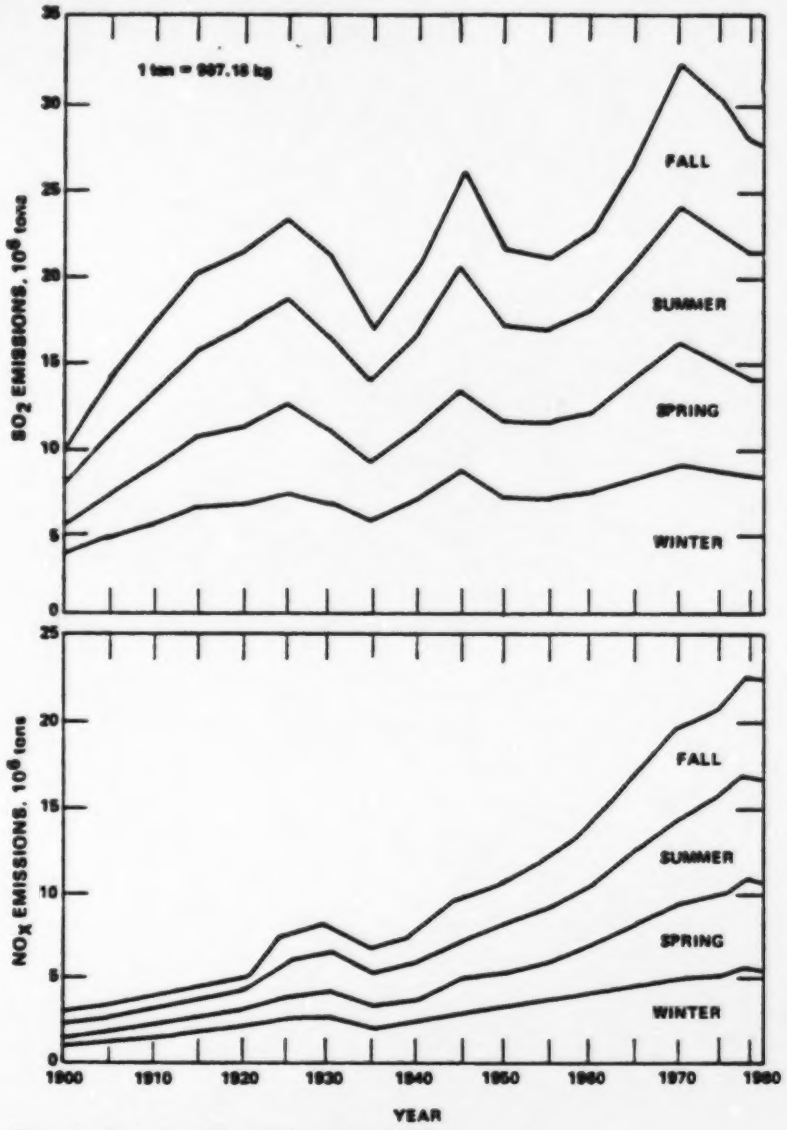

Figure 11. Total national SO_2 and NO_x emissions by season.

tities and types of fuels used and the types of combustion sources. To varying degrees, the trend in emissions has been affected by a growth in population, the level of industrialization, the availability of fuels, and changes in the use of fuels. The trend of SO_2 emissions has also been affected by trends in the sulfur content of coals and oils consumed, especially during the past decade.

When comparing the trend of one state to another or one region to another, it must be remembered that the emission estimates do not represent emission density. (Emission density refers to the total state emissions divided by the area of the state.) Large states may have the highest total emissions, but this does not necessarily mean that smaller states do not have the same or even higher emission densities. For example, Illinois, Ohio, Pennsylvania, and New York have historically had the greatest SO_2 emissions of all states, but at various times their emission densities were comparable to those of many other smaller states such as Delaware, New Jersey, and Kentucky.

The historic emission estimates derived by this study are consistent in methodology from state to state and from year to year. These state-level estimates may provide sufficient spatial resolution to determine a general relationship between emissions and observed environmental effects. The estimates provide a basis for studies of the effects presently underway as part of the National Acid Precipitation Assessment Program. In other studies, Hidy *et al.* investigated the relationship between the SO_2 emission trends on a regional scale and historic deposition monitoring data.[27,28] In another study, Smith and Alexander investigated the relationship between historic emissions and stream chemistry data collected at various sites throughout the country.[29] The historic emission trends provide background information useful in formulating public policy regarding acid rain.[30] The present emission data file represents the best estimates to date on the state level. As research continues, further refinements in the methodology can be made.

Further improvements would involve completing the data bases on state fuel consumption and sulfur content values for the earlier years and development of historic emission factors. However, the greatest obstacle to further improvement is the lack of available information. Based on the findings of an extensive literature search conducted during this study, it is unlikely that any additional state-specific data can be obtained. The completion of the data base will require making additional assumptions as described by Gschwandtner *et al.* regarding unavailable data.[10]

The aggregated state emissions derived by this study were compared to the national total emissions reported by EPA[4] and the total state emissions of electric utilities estimated by Pechan and Wilson.[31,32] The comparisons indicate agreement with these other estimates for all years in common. The aggregated state totals are within 5.8 percent of the total national emissions alone from 1940 to 1980 estimated by EPA. The aggregated emissions of electric utilities are within 8.4 percent of the estimates by Pechan and Wilson. These differences are due to inherent differences in methodology, calculation procedures, and numerical rounding. The close agreement with more detailed studies adds some reassurance to the historical estimates. In a recent independent study, Husar estimated the SO_2 and NO_x emission trend in the eastern United States.[33] The aggregated emissions of both studies show a similar trend.

Uncertainties and Limitations

The greatest limitation to making historic emission estimates is the unavailability of information for certain years and source categories. Uncertainty is introduced by the need to make assumptions in lieu of missing data. It is also introduced by the methodology and, to a lesser extent, by the calculation procedure.

The greatest potential uncertainty concerns the use of time-invariant emission factors. For this study, special emission factors were developed which

represent each source category. This required weighting AP-42 emission factors according to the quantity of fuel consumed by each individual type of boiler, furnace, process, or engine contained within the category, and obtaining an average factor. The historic distribution of fuel to individual type of source is poorly known. It is generally known only for broad categories. As a result, the emission factors were held constant over time for most categories. This may be a particular limitation of the NO_x emission factors prior to 1960, when it is commonly believed that combustion processes were less carefully controlled and combustion temperatures were generally lower than today. Further research will be needed before emission factors can be adjusted for specific time periods.

Uncertainty is also introduced by applying sulfur content values for 1955 to earlier years. This assumption appears reasonable considering that the quality of coal as mined (the major fuel in the early years) appears to have remained fairly constant for major coal producing districts. However, the distribution of coal from mine to consuming state has changed over the study period. Data on coal distribution from producing district to end-use are not generally available prior to 1950, in a consistent manner.

Often it was necessary to extract state level data from larger scale data and to segregate fuel consumption according to each source category. These procedures were necessary for many of the earlier years and may introduce additional error in the emission estimates.

Estimates of the probable errors associated with the total state emissions can be based on the results of the uncertainty analysis presently being planned for the NAPAP 1980 emission inventory.[34] The percentage difference between the NAPAP 1980 and the historic emission estimates for all states combined is only 5.9 percent. When the uncertainty analysis of the NAPAP emission inventory is completed, it can serve as the benchmark from which the probable errors of historic estimates can be interpreted. Error bounds associated with each assumption will need to be established. As is often the case with historical work, further estimation will require making additional assumptions.

Conclusion

The current historic emissions data file presents the best estimates available on the state level. The data file is consistent in methodology and complete for the time period from 1950 to 1980. For earlier years, reasonable estimates are available. The emission trends of each state vary over time reflecting changes in a variety of economic and technological factors. While the national and regional scale emissions data provide general indications of trends, it is recommended that the reader refer to the state-specific estimates available for studies of the historical relationships between emissions and environmental effects. These emission estimates can serve as the basis for future studies of the relationship between emissions and environmental effects associated with the acidic deposition phenomenon.

Acknowledgment

This project was supported by funding from Task Group B, Man-Made Sources, National Acid Precipitation Assessment Program, through the U.S. EPA's Air and Energy Engineering Research Laboratory.

The authors are grateful to Rudolf B. Husar and to Frederick Lipfert for sharing information with us and for consultation and valuable suggestions. The authors are also indebted to many individuals in many agencies who provided copies of relevant historical data and to the staff of Duke University Library's Reference Department for assistance in locating information. Suggestions and comments of the technical reviewers of the draft document are appreciated and hereby acknowledged.

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